

FORM PTO-1396 (Modified)  
(REV 11-98)

U.S. DEPARTMENT OF COMMERCE PATENT AND TRADEMARK OFFICE

ATTORNEY'S DOCKET NUMBER

TRANSMITTAL LETTER TO THE UNITED STATES  
DESIGNATED/ELECTED OFFICE (DO/EO/US)  
CONCERNING A FILING UNDER 35 U.S.C. 371

37332-151134

U.S. APPLICATION NO. (IF KNOWN, SEE 37 CFR

09/856323

INTERNATIONAL APPLICATION NO.

PCT/KR99/00524

INTERNATIONAL FILING DATE

September 7, 1999

PRIORITY DATE CLAIMED

November 19, 1998

TITLE OF INVENTION

Miniature Fuel Reformer And System Using Metal Thin Film

APPLICANT(S) FOR DO/EO/US

Jaesung Han, Cheolwoo Park, Keunseob Choi, Ilsu Kim

Applicant herewith submits to the United States Designated/Elected Office (DO/EO/US) the following items and other information:

1. ☒ This is a **FIRST** submission of items concerning a filing under 35 U.S.C. 371.
2. ☐ This is a **SECOND** or **SUBSEQUENT** submission of items concerning a filing under 35 U.S.C. 371.
3. ☒ This is an express request to begin national examination procedures (35 U.S.C. 371(f)) at any time rather than delay examination until the expiration of the applicable time limit set in 35 U.S.C. 371(b) and PCT Articles 22 and 39(1).
4. ☒ A proper Demand for International Preliminary Examination was made by the 19th month from the earliest claimed priority date.
5. ☒ A copy of the International Application as filed (35 U.S.C. 371 (c) (2))
  - a. ☐ is transmitted herewith (required only if not transmitted by the International Bureau).
  - b. ☒ has been transmitted by the International Bureau.
  - c. ☐ is not required, as the application was filed in the United States Receiving Office (RO/US).
6. ☐ A translation of the International Application into English (35 U.S.C. 371(c)(2)).
7. ☒ A copy of the International Search Report (PCT/ISA/210).
8. ☒ Amendments to the claims of the International Application under PCT Article 19 (35 U.S.C. 371 (c)(3))
  - a. ☐ are transmitted herewith (required only if not transmitted by the International Bureau).
  - b. ☐ have been transmitted by the International Bureau.
  - c. ☐ have not been made; however, the time limit for making such amendments has NOT expired.
  - d. ☒ have not been made and will not be made.
9. ☐ A translation of the amendments to the claims under PCT Article 19 (35 U.S.C. 371(c)(3)).
10. ☐ An oath or declaration of the inventor(s) (35 U.S.C. 371 (c)(4)).
11. ☒ A copy of the International Preliminary Examination Report (PCT/IPEA/409).
12. ☐ A translation of the annexes to the International Preliminary Examination Report under PCT Article 36 (35 U.S.C. 371 (c)(5)).

## Items 13 to 20 below concern document(s) or information included:

13. ☐ An Information Disclosure Statement under 37 CFR 1.97 and 1.98.
14. ☒ An assignment document for recording. A separate cover sheet in compliance with 37 CFR 3.28 and 3.31 is included.
15. ☐ A **FIRST** preliminary amendment.
16. ☐ A **SECOND** or **SUBSEQUENT** preliminary amendment.
17. ☐ A substitute specification.
18. ☒ A change of power of attorney and/or address letter.
19. ☒ Certificate of Mailing by Express Mail
20. ☒ Other items or information:

Express Mail Label No. EL 813775324 US



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PATENT TRADEMARK OFFICE

U.S. APPLICATION NO. (IF KNOWN, SEE 37 CFR 1.53) <b>09/856323</b>	INTERNATIONAL APPLICATION NO. <b>PCT/KR99/00524</b>	ATTORNEY'S DOCKET NUMBER <b>37332-151134</b>
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21. The following fees are submitted: <b>BASIC NATIONAL FEE (37 CFR 1.492 (a) (1) - (5)):</b> <input checked="" type="checkbox"/> Neither international preliminary examination fee (37 CFR 1.482) nor international search fee (37 CFR 1.445(a)(2)) paid to USPTO and International Search Report not prepared by the EPO or JPO ..... <b>\$1,000.00</b> <input type="checkbox"/> International preliminary examination fee (37 CFR 1.482) not paid to USPTO but International Search Report prepared by the EPO or JPO ..... <b>\$860.00</b> <input type="checkbox"/> International preliminary examination fee (37 CFR 1.482) not paid to USPTO but international search fee (37 CFR 1.445(a)(2)) paid to USPTO ..... <b>\$710.00</b> <input type="checkbox"/> International preliminary examination fee paid to USPTO (37 CFR 1.482) but all claims did not satisfy provisions of PCT Article 33(1)-(4) ..... <b>\$690.00</b> <input type="checkbox"/> International preliminary examination fee paid to USPTO (37 CFR 1.482) and all claims satisfy provisions of PCT Article 33(1)-(4) ..... <b>\$100.00</b>	<b>CALCULATIONS PTO USE ONLY</b>															
<b>ENTER APPROPRIATE BASIC FEE AMOUNT =</b>	<b>\$1,000.00</b>															
Surcharge of <b>\$130.00</b> for furnishing the oath or declaration later than <input type="checkbox"/> 20 <input type="checkbox"/> 30 months from the earliest claimed priority date (37 CFR 1.492 (e)).																
<table border="1" style="width: 100%; border-collapse: collapse;"> <tr> <th style="width: 20%;">CLAIMS</th> <th style="width: 20%;">NUMBER FILED</th> <th style="width: 20%;">NUMBER EXTRA</th> <th style="width: 20%;">RATE</th> <th style="width: 20%;"></th> </tr> <tr> <td>Total claims</td> <td>8 - 20 =</td> <td>0</td> <td>x \$18.00</td> <td style="text-align: right;"><b>\$0.00</b></td> </tr> <tr> <td>Independent claims</td> <td>2 - 3 =</td> <td>0</td> <td>x \$80.00</td> <td style="text-align: right;"><b>\$0.00</b></td> </tr> </table>	CLAIMS	NUMBER FILED	NUMBER EXTRA	RATE		Total claims	8 - 20 =	0	x \$18.00	<b>\$0.00</b>	Independent claims	2 - 3 =	0	x \$80.00	<b>\$0.00</b>	<b>\$0.00</b>
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Fee for recording the enclosed assignment (37 CFR 1.21(h)). The assignment must be accompanied by an appropriate cover sheet (37 CFR 3.28, 3.31) (check if applicable). <input checked="" type="checkbox"/>																
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☒ A check in the amount of **\$1,040.00** to cover the above fees is enclosed.


☐ Please charge my Deposit Account No. \_\_\_\_\_ in the amount of \_\_\_\_\_ to cover the above fees.  
 A duplicate copy of this sheet is enclosed.

☒ The Commissioner is hereby authorized to charge any fees which may be required, or credit any overpayment to Deposit Account No. **50-0573** A duplicate copy of this sheet is enclosed.

**NOTE: Where an appropriate time limit under 37 CFR 1.494 or 1.495 has not been met, a petition to revive (37 CFR 1.137(a) or (b)) must be filed and granted to restore the application to pending status.**

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**30,480**  
 REGISTRATION NUMBER  


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**May 18, 2001**  
 DATE  


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PCT/KR99/00524

MINIATURE FUEL REFORMER AND SYSTEM USING METAL THIN FILM

BACKGROUND OF THE INVENTION

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Field of the Invention

The present invention relates in general to a fuel reformer utilizing a metal thin film for use in a fuel cell driven mobile vehicle which makes use of, as a fuel, alcohol such as methanol, ethanol, etc. More particularly, the invention relates to a miniature fuel reformer and a system thereof for use in a fuel cell driven mobile vehicle, which enables a reduction in the entire size by adopting an integrated hydrogen generation/separation type in which the steam reforming reaction for the hydrogen generation, and the separation reaction for the hydrogen separation are carried out simultaneously in the same reactor by the metal film property which selectively permeates only hydrogen of a reforming gas containing hydrogen, and by the principle by which the reaction rate increases beyond its equilibrium limit.

Description of the Prior Art

In general, a fuel cell system by which the chemical energy converts directly to the electric energy with a chemical reaction between a hydrogen gas and an electrolyte, is used in the field in which no internal combustion engine is substantially utilized, such as in the case of spaceship. The fuel cell system is also used as a system for supplying electric power to an

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electric driven mobile vehicle. In particular, in recent years, the importance of the fuel cell system is increasingly highlighted due to the tightening of restrictions on air pollution, and the limitation of the development of electric vehicles using battery power.

Moreover, with increasing interest in energy efficiency and the environmental pollution problem, it is accepted that internal engine driven vehicles can be virtually replaced with electric vehicles using fuel cells. However, there are various technically limiting factors that need still to be resolved, such that these fuel cell driven vehicles be widely commercially available. In particular, in a fuel cell driven vehicle that uses hydrogen gas as a fuel, all the problems with the construction of infrastructure for the storage, delivery, and supply of the hydrogen fuel act as limiting factors in virtually widely utilizing the fuel cell driven vehicle.

Thus, the development of a fuel reformer, wherein a liquid fuel such as methanol, ethanol, gasoline, and the like is reformed to generate hydrogen which is then separated for utilization as a fuel, has made recent progress. However, in order for such a fuel reformer to be widely utilized, developing a fuel reformer having smaller size, lighter weight, and excellent response to load is necessary. Particularly significant is that this fuel reformer is made smaller and lighter so that it can be mounted on the vehicle.

In addition, in recent fuel cell vehicles, there is used a Solid Polymer Electrolyte Fuel Cell(hereinafter, called "SPEFC"). Such a SPEFC, however, has a shortcoming

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in that it loses easily its activity by virtue of CO contained in hydrogen gas, that is, the fuel. For this reason, it is significant to lower the concentration of CO to a minimum.

5 Furthermore, to develop a fuel cell vehicle using a liquid fuel, thereby resolving the above described problems with respect to the use of the hydrogen gas fuel, a process for preparation of hydrogen that utilizes the prior art steam reforming reaction is developed  
10 worldwide. However, this technology has a problem in decreasing the total size of the fuel reformer system, because the steam reforming reaction and the hydrogen separation reaction are carried out in different reactors as shown in FIG. 1.

15 For example, in US Patent No. 4,613,436, there is an effort to decrease the size of a hydrogen separation apparatus by arranging a plurality of vertically spaced circular hydrogen separation membranes while interposing airtight protrusions between adjacent ones of the  
20 membranes such that spaces each defined between adjacent membranes by the airtight protrusions are isolated from one another. Through each of the hydrogen separation membranes is passed the reforming gas flow to separate hydrogen which is then captured. However, the structure  
25 of this hydrogen separation apparatus is very unsuitable for mass production, and also has difficulty in maintaining the airtightness at fixed portions. To resolve these problems, US Patent No. 5,536,405 proposes inserting gaskets between the hydrogen separation  
30 membranes during the arrangement of the membranes such that the resultant apparatus may be suitable for mass

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production and may also easily maintain the airtightness between the hydrogen separation membranes. A drawback with the apparatus in the latter patent, however, is that it is heavy in weight due to the application of the gasket means.

Meanwhile, an attempt to improve the performance of the fuel reformer was made. US Patent No. 5,458,857 attempted to carry out, in separate regions, the hydrogen gas generation process by the endothermic steam reforming of methane, and the exothermic transition reaction for transition of carbon monoxide and steam to carbon dioxide and hydrogen so as to improve the efficiency of the fuel reformer, thereby realizing the miniaturization of the fuel reformer. However, the problems associated with this patent are that the necessary heat cannot be supplied by only the heat from the transition reaction, and the structure of the fuel reformer used is complicated such that the efficiency is virtually lowered. Furthermore, US Patent No. 5,741,474 attempted to simultaneously practice, in a certain space, the partial oxidation of methane, as an exothermic reaction, and the reforming reaction, as an endothermic reaction, thereby maximizing the heat transfer efficiency. However, there is a drawback in that both reactions cannot be efficiently controlled, thereby to reduce the performance of the fuel reformer. Japanese Patent Application No. 93-147902 attempted to supply the necessary heat for the reaction by re-using, as a fuel, some of the hydrogen generated by the reaction of methanol with water, so as to improve the performance of the fuel reformer and to miniaturize the fuel reformer as well. However, there is a problem in

The above described inventions have limitations on the performance improvement and the miniaturization of the fuel reformer, because they attempted the performance improvement and the miniaturization by the improvement of the structure of the fuel reformer, without resolving the prior art problem in that the fuel reformer and the hydrogen separation apparatus must be separately present.

In an attempt to overcome such limitations, US Patent No.5,888,273 discloses a hydrogen generation device in which, after positioning a long tube-shaped membrane for separation of hydrogen and a steam reforming catalyst in the same space, hydrogen gas is separated, and some of the reforming gases, containing hydrogen, carbon monoxide, carbon dioxide, steam, etc. are transported to a separate location, and then are burned, thereby supplying the necessary heat for the reforming. However, such a device is problematic in that an initial ignition is difficult. Another problem is that the heat transfer efficiency of the device in supplying the necessary initial heat for the start of the reforming reaction, and heat for maintaining a necessary membrane temperature for the permeation of hydrogen, is poor. These problems are considered the significant drawbacks against the fast initial starting period and dynamic response of the fuel cell driven vehicle. In addition, because of the complicated structure and the use of the long tube-shaped hydrogen separation membrane, this

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hydrogen generation device is unsuitable for a process, for example a manufacturing process of vehicles, for which mass production is required.

5 SUMMARY OF THE INVENTION

It is therefore an object of the present invention to provide a miniature fuel reformer in which a metal thin film selectively permeating only hydrogen from a  
10 fuel is utilized to lower, to a very low degree, a concentration of carbon monoxide(CO) contained in the hydrogen fuel supplied to an anode of a SPEFC, thereby capable of reducing remarkably a poisoning of the anode by carbon monoxide.

15 It is other object of the present invention to provide a miniature fuel reformer in which unit modules are connected with each other in series, parallel, or a combination thereof, thereby capable of supplying, in a suitable way, a necessary amount of hydrogen to a  
20 vehicle.

It is another object of the present invention to provide a unit module in which a reaction for reforming of a liquid fuel and a reaction for separation of hydrogen are carried out in the same reactor as shown in  
25 FIG. 2, and therefore to provide a miniaturized fuel reforming system that can be easily mounted within the fuel cell driven vehicle.

To accomplish the above objects, the present invention provides a miniature fuel reformer for use in  
30 a fuel cell driven vehicle comprising a plurality of unit modules connected with each other in series, parallel, or

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- a combination thereof, each of the unit modules including; an inner housing having a cylindrical shape while having top and bottom walls; an outer housing arranged around the inner housing while being radially spaced from the inner housing; a hydrogen separation reaction chamber defined in the inner housing;
- 5 a combustion catalyst chamber defined between the inner and outer housings and contained with a combustion catalyst for supplying heat needed in the inner housing;
- 10 a fuel inlet provided at the top wall of the inner housing and adapted to introduce liquid fuel into the hydrogen separation reaction chamber;
- a plurality of vertically spaced hydrogen gas separation cells arranged in the hydrogen separation reaction chamber and adapted to selectively permeate hydrogen
- 15 contained in the fuel introduced in the hydrogen separation reaction chamber, thereby separating the hydrogen from the fuel; a steam reforming catalyst filled in the hydrogen separation reaction chamber between
- 20 adjacent ones of the hydrogen gas separation cells and adapted to reform the fuel introduced in the hydrogen separation reaction chamber; a plurality of vertically spaced cylindrical support members centrally arranged in the hydrogen separation reaction chamber while defining
- 25 a hydrogen passage communicating with the hydrogen gas separation cells, thereby allowing the hydrogen separated from the fuel to be discharged from the hydrogen gas separation cells, each of the cylindrical support members being interposed between adjacent ones of the hydrogen
- 30 gas separation cells and serving to regulate a gap defined between the adjacent hydrogen gas separation

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cells; a permeate discharge tube coupled to the bottom wall of the inner housing, the permeate discharge tube communicating with the hydrogen passage defined by the cylindrical support members to allow the hydrogen introduced in the hydrogen passage to be outwardly discharged from the hydrogen passage; a plurality of raffinate outlets provided at the bottom wall of the inner housing and adapted to discharge a portion of the fuel, not permeating any one of the hydrogen gas separation cells, from the hydrogen separation reaction chamber; and a plurality of combustion fuel/air inlets respectively connected to the raffinate outlets by conduits and adapted to introduce the fuel portion discharged from the raffinate outlets into the combustion catalyst chamber, along with air, so that the fuel portion is burned in the combustion catalyst chamber.

Moreover, the present invention provides a miniature fuel reforming system for use in fuel cell driven vehicles, comprising: a fuel preheater for preheating and vaporizing a liquid fuel; a fuel reformer for receiving the vaporized fuel via a fuel supply header and separating hydrogen from the received fuel to produce a reforming gas, the fuel reformer having a hydrogen separation reaction chamber for separating hydrogen from the received fuel, and a combustion catalyst chamber containing a combustion catalyst for supplying heat needed in the hydrogen separation reaction chamber, the combustion catalyst chamber receiving the hydrogen-separated fuel, that is, a raffinate; means for receiving a permeate, that is, the hydrogen separated by and discharged from the hydrogen separation reaction chamber

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of the fuel reformer, via a permeate header, controlling the pressure of the received permeate, and supplying the pressure-controlled permeate to an anode of a fuel cell; means for controlling the pressure of the raffinate discharged from the hydrogen separation reaction chamber, thereby controlling the hydrogen separation reaction chamber of the fuel reformer; and valve means for supplying the raffinate to the combustion catalyst chamber of the fuel reformer.

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## BRIEF DESCRIPTION OF THE DRAWINGS

The foregoing and other objects, features and advantages of the invention will be apparent to those skilled in the art to which the present invention relates from reading the following specification with reference to the accompanying drawings, in which:

FIG. 1 is a block diagram schematically showing that the steam reforming reaction and the hydrogen separation reaction are separately carried out with the prior art fuel reforming system.

FIG. 2 is a block diagram schematically showing that the steam reforming reaction and the hydrogen separation reaction are carried out in the same reactor with a fuel reformer according to the present invention.

FIG. 3 is a schematic view of the construction of a unit module of a fuel reformer according to the invention.

FIG. 4 is a schematic view of a construction of a hydrogen gas separation cell according to the invention, that permeates hydrogen from a reformed gas containing

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hydrogen.

FIG. 5 and FIG. 6 each is a schematic view showing passages for a fuel for the steam reforming, and for a permeate that is hydrogen permeated through the hydrogen gas separation cell, and also for a raffinate that is hydrogen non-permeated through the cell.

#### DESCRIPTION OF THE PREFERRED EMBODIMENTS

FIG. 3 schematically shows the construction of a unit module 1 included in a miniature fuel reformer for a fuel cell driven vehicle according to the invention.

A plurality of the unit modules 1 are connected in series, parallel, or a combination thereof with each other, to constitute the fuel reformer of the invention. The unit module 1 has a fuel inlet 17 which is formed on its upper end and into which a liquid fuel, for example, alcohol such as methanol, ethanol, etc., is introduced. Moreover, the unit module 1 has a hydrogen separation reaction chamber 7. In the hydrogen separation reaction chamber 7, a steam reforming catalyst 3 is contained which serves to reform the fuel introduced through the fuel inlet 17. The hydrogen separation reaction chamber 7 also contains therein a plurality of vertically spaced hydrogen gas separation cells 4 for permeating and separating only hydrogen from the reforming gas. The hydrogen separation reaction chamber 7 is defined in an inner housing 8 having a cylindrical shape while having top and bottom walls. Between adjacent ones of the hydrogen gas separation cells 4, there is interposed a plurality of vertically spaced cylindrical support

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members 6 fixed to those cells 4 at both ends thereof in accordance with a diffusion welding process. These cylindrical support members 6 are centrally arranged in the hydrogen separation chamber 7 while defining a hydrogen passage communicating with the hydrogen separation cells 4, thereby allowing the hydrogen separated from the fuel to be discharged from the hydrogen gas separation cells 4. The cylindrical support members 6 also serve to control a gap defined between the adjacent cells 4.

A combustion catalyst chamber 9 is arranged around the inner housing 8. The combustion catalyst chamber 9 has a combustion catalyst 5 for transferring the necessary heat to the hydrogen separation reaction chamber 7. This combustion catalyst chamber 9 is defined between the inner housing 8 and an outer housing 10 arranged around the inner housing 8.

A permeate discharge tube 25 is coupled to the bottom wall 1 of the inner housing 8. This permeate discharge tube 25 is also communicated with the hydrogen passage defined by the cylindrical support members 6 to allow the hydrogen introduced into the hydrogen passage to be outwardly discharged from the hydrogen passage. Meanwhile, in order for a raffinate, that is, a reforming gas non-permeated through the hydrogen gas separation cells, to discharge from the hydrogen separation reaction chamber, a plurality of raffinate outlets 25 are formed at an inclined bottom wall of the inner housing 8. In addition, although not shown in FIG. 3, a plurality of combustion fuel/air inlets respectively connected to the raffinate outlets by conduits. These combustion fuel/air

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inlets are adapted to introduce the fuel portion discharged from the raffinate outlets 25 into the combustion catalyst chamber 9, along with air, so that the fuel portion is burned in the combustion catalyst chamber 9.

Meanwhile, the hydrogen gas separation cells 4 each includes one or more channels 13 at its peripheral portion to allow the fuel to flow therethrough.

The construction of each of the hydrogen gas separation cells will now be described in detail with reference to FIG.4. Each hydrogen gas separation cell 4 comprises a pair of spaced metal films 11 and 11a adapted to selectively separate only hydrogen, and an annular support plate 12 made of copper or a copper alloy and provided at its peripheral portion with one or more vertical fuel channels 13. The support plate 12 is attached to the metal films 11 and 11a at opposite surfaces thereof, and therefore supports the metal thin film 11 and 11a. The hydrogen gas separation cell 4 also includes a support layer 14 (a porous separation membrane) serving to provide a passage for hydrogen permeated through the metal film 11 while maintaining a structural stability of the hydrogen gas separation cells 4. There is also a central support disk 15 made of copper or a copper alloy. The central support disk 15 is provided with a central hole denoted by no reference numeral and at least one radial hole 16. The radial hole 16 extends radially throughout the diameter of the central support disk 15 while communicating with the central hole formed at the central support disk 15 such a fashion that permeate, that is, hydrogen permeated

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through the metal films 11 and 11a, passes therethrough. Also, the central hole is communicated with the hydrogen passage defined by the cylindrical support members 6.

Moreover, in order to prevent a degradation in the hydrogen separation performance of the steam reforming catalyst 3 by a direct contact of the steam reforming catalyst 3 with the metal film 11, a layer, which may be the support layer 14, is disposed between the steam reforming catalyst 3 and the metal film 11. Preferably, the support layer 14 is made of a porous separation membrane.

In such a hydrogen gas separation cell 4, hydrogen in the reforming gas generated by reforming the fuel with the steam reforming catalyst 3 is adsorbed onto an outer surface of the metal film 11 and separated from the fuel. The separated hydrogen atoms are permeated through the metal film 11 and then are bonded into hydrogen molecules at an inner surface of the metal film 11. The hydrogen molecules are passed through the radial hole and central hole formed at the central support disk 16 and then discharged from the hydrogen separation reaction chamber 7 through the permeate discharge tube 25.

In such a hydrogen selective-separation reaction, a higher pressure differential between the inner surface and the outer surface of the metal film 11, as well as a higher surface temperature, result in an increased permeation performance of the metal film 11. In addition, to keep the surface of the metal film 11 clean is necessary for better permeation performance of the metal film. The metal films 11 and 11a used in the invention includes the film made of palladium-based alloy, such as

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Pd-Cu based alloy or Pd-Ag based alloy, and a metal film or metal foil made of vanadium(V), nickel(N) or an alloy thereof and coated with palladium.

Moreover, the steam reforming catalyst 3 useful in  
5 the invention includes a steam reforming catalyst based on Cu-Zn alloys, as well as a steam reforming catalyst containing precious metals in the amount of at least 0.01% by weight, such as Pt.

Furthermore, a hydrogen production capacity of the  
10 respective unit modules 1 in the fuel reformer can be varied freely depending on the surface area of the respective metal film 11, and the amount of the steam reforming catalyst 3 being filled. This allows the capacity of the modules to regulate freely. Namely, the  
15 amount of the steam reforming catalyst 3 filled can be varied depending on a height of the respective cylindrical support members 6, and a surface area of the metal film 11 can be varied with the diameter of the support plate 12. Accordingly, the hydrogen production  
20 capacity of the respective unit modules in the fuel reformer can be determined freely depending on the surface area of the respective metal film 11, and the amount of the steam reforming catalyst 3.

The support plate 12 supporting the metal films 11  
25 and 11a, and the central support disk 15, are connected with each other by diffusion welding such that there is no gap between the inside and the outside of the respective hydrogen gas separation cells 4. Therefore, the concentration of other reforming gases poisonous to  
30 the fuel cell stack, particularly carbon monoxide, is very low, thereby maintaining the performance of the fuel

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cell stack for a longer time.

As described above, it is preferable that the fuel channels 13 formed at the peripheral portion of the support plate 12 are arranged in a zig-zag as shown in FIG. 3, when connecting the respective hydrogen gas separation cells 4 with the cylindrical support members 6. This provides an increase in the contact efficiency between the fuel and the steam reforming catalyst 3, as well as an increase in the contact efficiency between the reforming gas and the metal film 11.

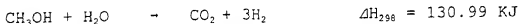
Referring to FIGS. 5 and 6, various passages and a reaction process will now be described below. In FIGS. 5 and 6, arrows shown as solid lines depict a fuel passage 19, arrows shown as dotted lines depict a permeate passage 18, and arrows shown as one dot chain lines depict a raffinate passage 20.

A fuel, for example, alcohol such as methanol, ethanol, etc., that is pre-vaporized by a fuel preheater(not shown) disposed outside of the fuel reformer, is introduced into the hydrogen separation reaction chamber 7 through the fuel inlet 16 formed at the top wall of the inner housing 8. The introduced fuel is heated with combustion heat from the combustion catalyst chamber 9 to a temperature in the range of from 150°C to 600°C that is the temperature required by the hydrogen separation reaction chamber 7. The heated fuel flows downwardly to the hydrogen gas separation cells 4 through the fuel channels 13 formed at the peripheral portion of the support plate 12 of the respective cells 4, while contacting with the steam reforming catalyst 3 filled therebetween. The fuel contacted with the catalyst

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3 is subjected to the reforming reaction as indicated in the following reaction scheme, to convert it into a reforming gas essentially consisting of hydrogen, carbon dioxide, and the remainder of water:

5



Such a reforming gas is then contacted with the metal film 11 that are connected to the support plate 12 by the diffusion welding. Only hydrogen of the fuel is selectively permeated through the metal films 11 and 11a, and is passed through the radial hole 16 formed at the central support disk 15, and then is discharged from the permeate discharge tube 25. Moreover, a raffinate that is a reformed gas non-permeated through the hydrogen gas separation cells 4 is discharged through raffinate outlets 26 formed along an inclined bottom wall of the inner housing 8, and is passed through conduits 29 and the combustion fuel/air inlets 21, and then introduced into the combustion catalyst chamber 9.

Meanwhile, the steam reforming reaction as indicated in the above reaction scheme is the endothermic reaction for which much heat is necessary. It is preferable that the necessary heat for such an endothermic steam reforming reaction is obtained by a heat exchange with the combustion heat. For such a heat exchange, methods may be used wherein a fuel stream supplied to the hydrogen separation chamber 7 and a fuel stream supplied to the combustion catalyst chamber 9 are the cocurrent flows, or wherein both streams are the countercurrent flows. However, the latter method is preferred for an

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increase in heat efficiency by more effective heat exchange. Namely, in the raffinate being discharged from the hydrogen separation chamber 7 without permeating the hydrogen gas separation cells 4, there is contained  
5 hydrogen in the range of 1 to 70% by weight that varies depending on operation conditions of the hydrogen separation reaction chamber 7. Accordingly, where the raffinate is used as a fuel in the combustion catalyst chamber 9, there is no need for the supply of a separate  
10 fuel. This can further improve the energy efficiency of the fuel reformer.

The above described unit modules are connected with each other in series, parallel, or a combination thereof to constitute the fuel reformer that is a part of the  
15 fuel reforming system.

Hereinafter, the operation of the fuel reforming system using the fuel reformer of the invention will be described with reference to FIG. 7.

The fuel reforming system according the invention  
20 comprises a fuel preheater 22 for preheating a liquid fuel, such as methanol, etc., a fuel reformer 30 in which a plurality of unit modules 1 are connected with each other in series, parallel, or a combination thereof, and a pressure controller 24 for controlling pressure of a  
25 permeate, that is, hydrogen discharged the unit modules. The fuel reforming system also includes a raffinate pressure controller 23 for controlling a pressure of hydrogen separation reaction chamber 7 of the respective unit modules 1, a fuel supply header 28 for supplying the  
30 liquid fuel to the respective unit modules 1, a permeate header 27 for collecting a permeate discharged from the

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hydrogen separation reaction chamber 7 of each unit module, and a valve 2 for supplying, to a combustion catalyst chamber of the respective unit modules, fuel non-permeated through metal thin films 11 in each unit module.

In such a fuel reforming system, the fuel, such as methanol, etc., is vaporized in the fuel preheater 22 comprising the fuel injector or the heat exchanger, and is passed to the fuel supply header 28. Thereafter, the fuel is passed through a fuel inlet 17 formed on the top wall of the inner housing arranged in the respective unit modules 1, and then introduced into the hydrogen separation reaction chamber 7 in which the fuel is subjected to the fuel reforming reaction and the hydrogen separation reaction simultaneously.

After the permeate generated by the above reactions is collected in the permeate header 27 again, it is supplied to an anode of the fuel cell stack. At this point, the pressure controlling device 24 composed of the compressor or the pressure controlling valve, etc. is disposed on the permeate header 27, such that, as occasion demands, the permeate pressure is controlled depending on the operating pressure of the fuel cell stack.

Moreover, the pressure of the hydrogen separation reaction chamber is controlled by the pressure controller, and the raffinate is passed through the pressure controller 23 to drop its pressure to an atmospheric pressure. Thereafter, the raffinate is passed to the valve 2. As occasion demands, the raffinate is passed from the valve 2 to the combustion catalyst

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chamber 9 to be utilized as the fuel. This highly increases the energy efficiency of the fuel reformer.

Furthermore, when the heat exchanger is used as the fuel preheater 22, the heat supplied to the fuel preheater is heat-exchanged with a combustion gas discharged from the combustion catalyst chamber 9, to increase the energy efficiency.

Additionally, as described above, the fuel reformer 30 has a construction capable of controlling the hydrogen production rate, in that the unit modules are simply connected with each other in series, parallel, or a combination thereof depending on the necessary hydrogen amount for the fuel cell stack. Thus, the fuel reformer is suitable for mass production.

Even though the fuel reformer of the invention corresponds to the hydrogen generation and supply apparatus, this may be also applied in various fields including devices that require the hydrogen generation and the hydrogen purification from the mixed gases containing hydrogen, particularly the production of hydrogen of high purity.

In particular, the fuel reformer of the invention utilizes the palladium-based metal film of the property selectively separating only hydrogen from the fuel to reduce remarkably the concentration of CO in a hydrogen fuel supplied to an anode of SPEFC. This provides reduction in the poisoning of the anode by CO, and also enables the production of the proper amount of hydrogen from the respective unit modules. In addition, the fuel reformer is essentially composed of a plurality of the unit modules connected simply with each other in series,

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parallel, or a combination thereof. The fuel reformer has a construction adapted for mass production, and therefore may supply various capacities of hydrogen required by the fuel cell driven vehicles.

5 When the inventive miniature fuel reformer using the metal thin film is applied to the fuel cell driven vehicle, the hydrogen production reaction (such as the steam reforming reaction) and the hydrogen separation reaction are carried out in the same reactor. As a  
10 result, the fuel reformer may supply, to an anode of the fuel cell, highly pure hydrogen having a remarkably low carbon monoxide concentration, without requiring a separate apparatus for hydrogen separation. This results in a decrease in poisoning of the anode by carbon  
15 monoxide. This also results in a remarkable decrease in size of the fuel reformer to allow the fuel reformer to easily mount in the vehicle. In addition to the above advantages, as the fuel reformer is fabricated with the unit module having certain hydrogen production capacity,  
20 to supply the varied amounts of hydrogen required by the fuel cell driven vehicle is possible with a simple module assembly only. This yields an efficient increase in the necessary assembly productivity for mass production.

25 Although the preferred embodiments of the invention have been disclosed for illustrative purposes, those skilled in the art will appreciate that various modifications, additions and substitutions are possible, without departing from the scope and spirit of the invention as disclosed in the accompanying claims.

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## WHAT IS CLAIMED IS:

1. A miniature fuel reformer for use in a fuel cell driven vehicle comprising a plurality of unit modules connected with each other in series, parallel, or a combination thereof, each of the unit modules including:

an inner housing having a cylindrical shape while having top and bottom walls;

10 an outer housing arranged around the inner housing while being radially spaced from the inner housing;

a hydrogen separation reaction chamber defined in the inner housing;

15 a combustion catalyst chamber defined between the inner and outer housings and containing a combustion catalyst for supplying heat needed in the inner housing;

20 a fuel inlet provided at the top wall of the inner housing and adapted to introduce liquid fuel into the hydrogen separation reaction chamber;

25 a plurality of vertically spaced hydrogen gas separation cells arranged in the hydrogen separation reaction chamber and adapted to selectively permeate hydrogen contained in the fuel introduced in the hydrogen separation reaction chamber, thereby separating the hydrogen from the fuel;

30 a steam reforming catalyst filled in the hydrogen separation reaction chamber between adjacent ones of the hydrogen gas separating cells and adapted to reform the fuel introduced in the

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hydrogen separation reaction chamber;

5 a plurality of vertically spaced cylindrical support members centrally arranged in the hydrogen separation reaction chamber while defining a hydrogen passage communicating with the hydrogen separating cells, thereby allowing the hydrogen separated from the fuel to be discharged from the hydrogen separation cells, each of the cylindrical support members being interposed between adjacent  
10 ones of the hydrogen gas separation cells and serving to regulate a gap defined between the adjacent hydrogen gas separation cells;

a permeate discharge tube coupled to the bottom wall of the inner housing, the permeate discharge  
15 tube communicating with the hydrogen passage defined by the cylindrical support members to allow the hydrogen introduced in the hydrogen passage to be outwardly discharged from the hydrogen passage;

a plurality of raffinate outlets provided at  
20 the bottom wall of the inner housing and adapted to discharge a portion of the fuel, not permeating through any one of the hydrogen gas separation cells, from the hydrogen separation reaction chamber; and

25 a plurality of combustion fuel/air inlets respectively connected to the raffinate outlets by conduits and adapted to introduce the fuel portion discharged from the raffinate outlets into the combustion catalyst chamber, along with air, so that  
30 the fuel portion is burned in the combustion catalyst chamber.

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2. The miniature fuel reformer of claim 1 wherein each of the hydrogen gas separation cells comprises:

a pair of spaced metal films adapted to selectively separate hydrogen from the fuel;

5 an annular support plate interposed between the metal films and attached to the metal films at opposite surfaces thereof, the support plate having at least one fuel channel at a portion thereof;

a support layer interposed between the metal films  
10 inside the support plate and adapted to provide a structural stability for the metal films to withstand a high pressure, the support layer having a porous separation film structure to provide a passage for the hydrogen introduced into the interior of the hydrogen gas  
15 separation cell defined between the metal films; and

a central support disk arranged inside the support layer in such a fashion that it is vertically aligned with the cylindrical support members, the central support disk having at least one radial hole communicating with  
20 the hydrogen passage provided by the support layer and with the hydrogen passage defined by the cylindrical support members, thereby serving as a permeate passage.

3. The miniature fuel reformer of claim 1 wherein  
25 the steam reforming catalyst contains at least one metal component selected from the group consisting of Cu, Zn, Fe, Cr, Ti, and Ni, or contains at least one precious metal in the amount of at least 0.01% by weight based on the weight of the catalyst.

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4. The miniature fuel reformer of claim 2 wherein

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the respective metal films is made of a palladium-based alloy or a metal foil coated with palladium.

5        5.    The miniature fuel reformer of claim 2 wherein  
the support plate and the metal films, and the central  
support disk and the metal films, respectively, are  
attached to each other by a diffusion welding.

10       6.    The miniature fuel reformer of claim 2 wherein  
the hydrogen gas separation cells are arranged in the  
hydrogen separation reaction chamber such that the fuel  
channels are located in a zigzag, whereby a contact  
efficiency between the fuel and the steam reforming  
catalyst, and a contact efficiency between the reforming  
15    gas and the metal films are increased.

20       7.    The miniature fuel reformer of claim 1 wherein  
the respective cylindrical support members is fixed to  
the hydrogen gas separation cells by a diffusion welding.

25       8.    A miniature fuel reforming system for use in  
fuel cell driven vehicles, comprising:  
a fuel preheater for preheating and vaporizing a  
liquid fuel; a fuel reformer for receiving the  
vaporized fuel via a fuel supply header and separating  
hydrogen from the received fuel to produce a reforming  
gas, the fuel reformer having a hydrogen separation  
reaction chamber for separating hydrogen from the  
received fuel, and a combustion catalyst chamber  
30    contained with a combustion catalyst for supplying heat  
needed in the hydrogen separation reaction chamber, the

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combustion catalyst chamber receiving the hydrogen-separated fuel, that is, a raffinate;

means for receiving a permeate, that is, the hydrogen separated by and discharged from the hydrogen separation reaction chamber of the fuel reformer, via a permeate header, controlling the pressure of the received permeate, and supplying the pressure-controlled permeate to an anode of a fuel cell;

means for controlling the pressure of the raffinate discharged from the hydrogen separation reaction chamber, thereby controlling the hydrogen separation reaction chamber of the fuel reformer; and valve means for supplying the raffinate to the combustion catalyst chamber of the fuel reformer.

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FIG 1

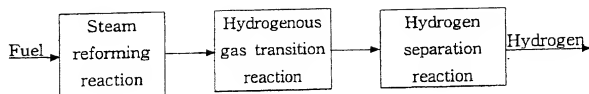
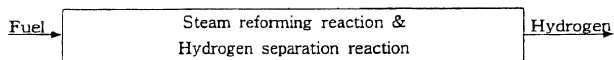
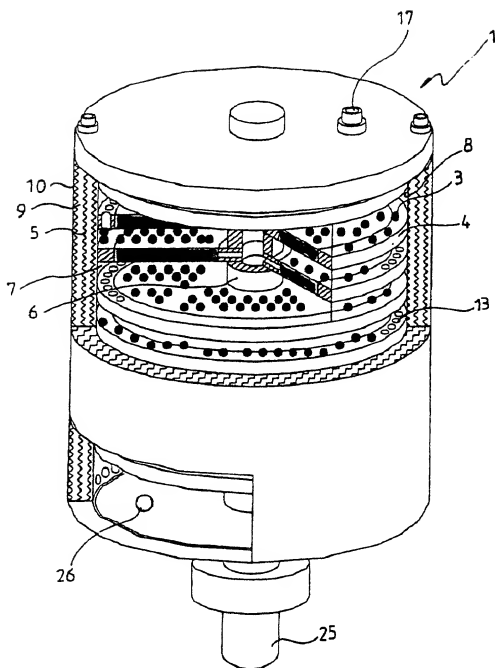


FIG 2



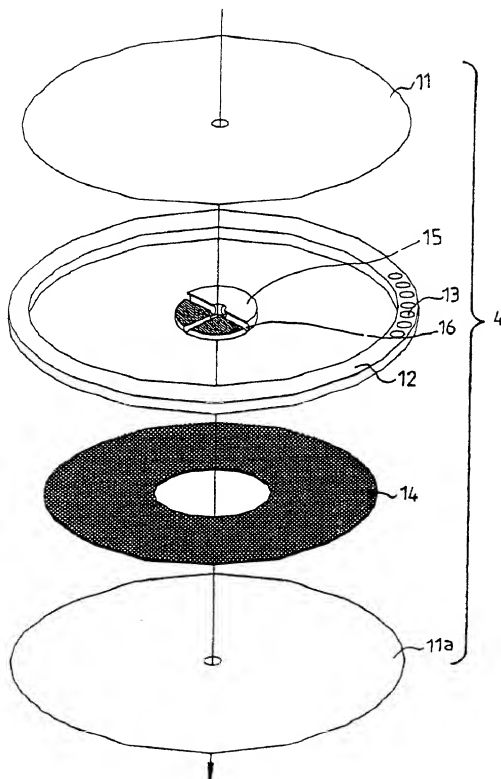
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FIG 3



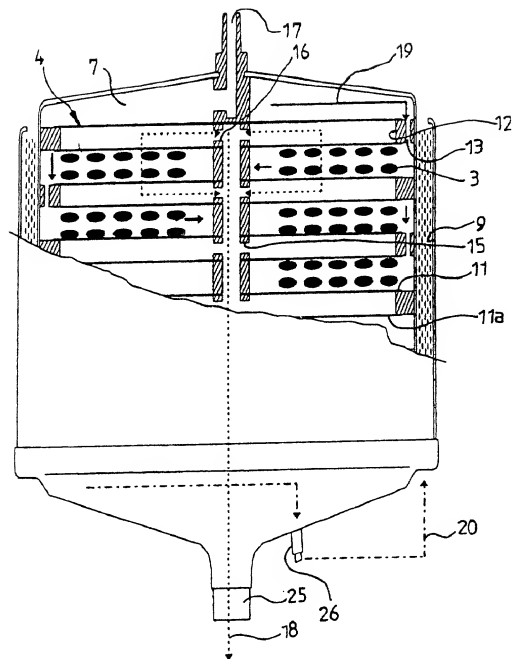
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FIG 4



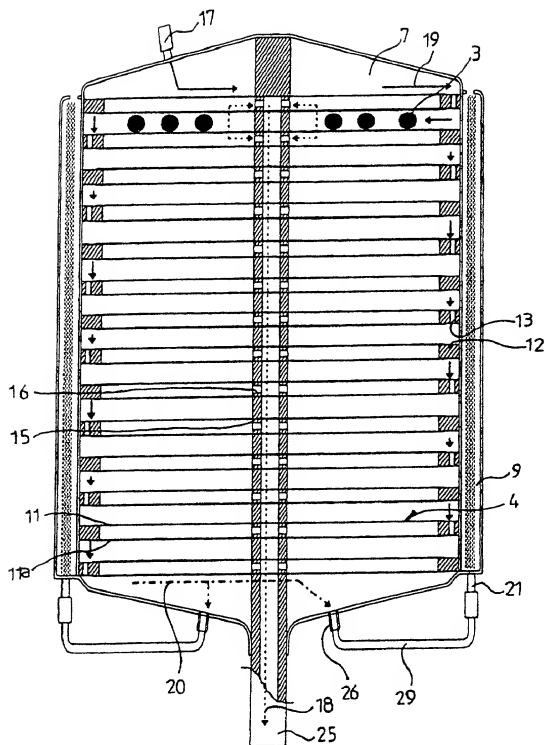
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FIG 5



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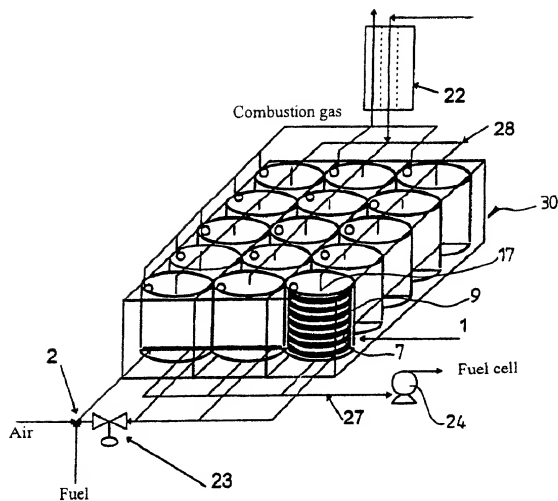
FIG 6





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FIG 7



ATTORNEY DOCKET #: \_\_\_\_\_

# DECLARATION AND POWER OF ATTORNEY

As a below named inventor, I hereby declare that:

My residence, post office address and citizenship are stated below next to my name.

I believe I am the original, first, and sole inventor (if only one name is listed below) or an original, first, and joint inventor (if plural names are listed below) of the subject matter which is claimed and for which a patent is sought on the invention entitled:

## TITLE OF INVENTION

MINIATURE FUEL REFORMER AND SYSTEM USING METAL THIN FILM

the specification of which is attached hereto unless the following box is checked

☒ was filed on \_\_\_\_\_ as Application Serial No. \_\_\_\_\_ or PCT Application No. PCT/KR99/00524 and was amended on \_\_\_\_\_ (if applicable).

I hereby state that I have reviewed and understand the contents of the above-identified specification, including the claims, as amended by any amendment referred to above.

I acknowledge the duty to disclose information which is material to the examination of this application in accordance with 37 CFR §1.56.

I hereby claim foreign priority benefits under 35 U.S.C. §119(a)-(d) or §365(b) of any foreign application(s) for patent or inventor's certificate, or §365(a) of any PCT international application which designated at least one country other than the United States, listed below and have also identified below any foreign application for patent or inventor's certificate or PCT International application having a filing date before that of the application on which priority is claimed:

### PRIOR FOREIGN/PCT APPLICATION(S)

COUNTRY	APPLICATION NO.	DATE OF FILING	PRIORITY CLAIMED
Korea	1998/049813	November 19, 1998	<input checked="" type="checkbox"/> YES NO <input type="checkbox"/>
			<input type="checkbox"/> YES NO <input type="checkbox"/>

I hereby claim the benefit under 35 U.S.C. §119(e) of any United States provisional application(s) listed below.

### PROVISIONAL APPLICATION NUMBER

### DATE OF FILING

I hereby claim the benefit under 35 U.S.C. §120 of any United States application(s) or §365(c) of any PCT International application(s) designating the United States, listed below and, insofar as the subject matter of each of the claims of this application is not disclosed in the prior United States application in the manner provided by the first paragraph of 35 U.S.C. §112, I acknowledge the duty to disclose material information as defined in 37 CFR §1.56 which became available between the filing date of the prior application and the national or PCT international filing date of this application:

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PRIOR U.S. APPLICATIONS OR PCT INTERNATIONAL APPLICATIONS  
DESIGNATING THE U.S. FOR BENEFIT UNDER 35 U.S.C. §120

Status (check one)

Application Serial No.	Date of Filing	Patented	Pending	Abandoned
		<input type="checkbox"/>	<input type="checkbox"/>	<input type="checkbox"/>
		<input type="checkbox"/>	<input type="checkbox"/>	<input type="checkbox"/>

And I hereby appoint Arthur H. Seidel, Registration No. 15,979; Gregory J. Lavorgna, Registration No. 30,469; Daniel A. Monaco, Registration No. 30,480; Thomas J. Durling, Registration No. 31,349; and John J. Marshall, Registration No. 29,671, my attorneys or agents with full power of substitution and revocation, to prosecute this application and to transact all business in the Patent and Trademark Office connected therewith.

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I hereby declare that all statements made herein of my own knowledge are true and that all statements made on information and belief are believed to be true; and further that these statements were made with the knowledge that willful false statements and the like so made are punishable by fine or imprisonment, or both, under Section 1001 of Title 18 of the United States Code, and that such willful false statements may jeopardize the validity of the application or any patent issuing thereon.

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